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Abstract: One of the requirements of the recently approved EU-BSS (European Union Basic Safety Standards) is the design and implementation of national radon action plans in the member states (Annex XVIII). Such plans require radon surveys. The analysis of indoor radon data is supported by the existing knowledge about geogenic radiation. With this aim, we used the terrestrial gamma dose rate data from the MARNA project. In addition, we considered other criterion regarding the surface of Spain, population, permeability of rocks, uranium and radium contain in soils because currently no data are available related to soil radon gas concentration and permeability in Spain. Given that, a Spanish radon map was produced which will be part of the European Indoor Radon Map and a component of the European Atlas of Natural Radiation. The map indicates geographical areas with high probability of finding high indoor radon concentrations. This information will support legislation regarding prevention of radon entry both in dwellings and workplaces. In addition, the map will serve as a tool for the development of strategies at all levels: individual dwellings, local, regional and national administration.

SPANISH EXPERIENCE ON THE DESIGN OF RADON SURVEYS BASED ON THE USE OF GEOGENIC INFORMATION

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Highlights

The last version of the Spanish indoor radon map accounts with 9,211 measurements.

Several criteria has been designed to plan a measurements campaign based on known correlations.

Lithostratigraphy is a good criterion for radon mapping (national scale). Local scales need others.

1. INTRODUCTION

The design of the Spanish indoor radon map is part of the framework of the European Commission's plan for the elaboration of the European Indoor Radon map, as a part of the European Natural Radiation Atlas (Dubois et al., 2010; Tollefsen et al., 2011; JRC_REM_EC, 2014; JRC_IES, 2012). This atlas will support policies in the field of public health, and it will contribute to increase the general public's awareness of the annual dose due to natural radioactivity.

A number of studies in various countries have proved that there is a clear correlation between exposure to radon inside buildings and the risk of developing lung cancer (ICRP 2010, 2011). Radon gas is responsible for between 3 and 4 per cent of deaths caused by this illness in the first world (IAEA, 2011; WHO, 2009), being the main source of ionizing radiation (EURATOM 1990, 1996, 2013; ICRP, 1994). Therefore, it is crucial to determine the areas where there is a greater probability of finding buildings with higher radon concentrations, as well as to analyse the variables which affect radon concentrations inside buildings.

Indoor radon concentration varies geographically. This is due to the large number of factors that affect radon appearance in buildings, such as the geology of the areas upon which buildings are constructed, soil permeability, specific rock characteristics, the meteorology and topography of the region, the proximity of active fault lines, the materials employed in construction, the design features of the buildings and the lifestyle habits of the occupants (García et al., 2013a). In our work we used a geographic information system (GIS) which enabled us to capture, store, create searches, analyse and visualize the statistical data we obtained. In order to adapt to the design of the European Indoor Radon map, Spain followed national (CSN 2012a; CSN 2012b) and international legislation (ICRP 1993, 2009, 2014).

This paper aims to produce a radon map of the Spanish territory that shows the probability of finding areas with levels of radon indoors, and is related to the European legislation that has to be implemented in the member states before the end of 2018 (EURATOM 2013). This map will be a very useful instrument for applying the requirements of European legislation on the radon issue at all administrative levels: national, regional and local.

2. METHODOLOGY

2.1 Definition of the grid

The grid was generated using the programme ArcGIS where the extreme corners are defined by established coordinates as such by the EC in the European Datum 1950 UTM Zone 30N (ED50) projection system: Peninsular Spain NW (433193,87 ; 4834808,73) SE (692906,43 ; 3961069,60) Canary Islands NW (692915,11 ; 3320469,29) SE (698454,23 ; 2988008,14). These coordinates were converted to longitude-latitude ED50 (decimal degrees) to generate a 10x10 km^2 grid of the Spanish surface in Google Earth format (.kml). The ED50 system is an old geodetic reference system used in Europe and has been used in coexistence with the ETRS89 in Spain until 2015. The parameters of this system are defined in the ArcGIS as shown in Table 1.

In order to follow a similar scheme as other EU member countries, we began working with a continental level projection system (GISCO-Lambert Acimutal Equal Area) following the guidelines established by the EC (Dubois et al., 2010; Tollefsen et al., 2014). The Lambert Azimuthal Equal Area projection is a planar projection, which means that map data are projected onto a flat surface.

In this way, measurements between countries can homogenized and the so call border effect avoided. Table 1 shows the parameters used to convert ED50 to GISCO-LAEA ArcGIS. As we show in Table 1, we took into account those parameters set by the EC. Therefore we obtain the following limits for 10x10 km^2 grid: Peninsular Spain NW (-1500000 ; -301000) SE (-351000 ; -1350000) Canary Islands NW (-2700000 ; -1900000) SE (-2150000 ; -1650000).

To define the working area, we used the administrative boundaries provided by the National Geographic Institute (IGN, 2014). Thus we generated a total number of 5,478 cells of 10x10 km^2 surface. For each cell, an identifying code was created and its centroid in meters (“x” and “y” coordinates) calculated. The statistical data obtained from the measurements were georeferenced to the centroid.

2.2 The measurement campaigns

The Spanish indoor radon map comprises, to date, 9,211 measurements, obtained over successive sampling campaigns (Figure 1). In each campaign, a series of measurements for each cell was defined, taking into account superficial, population, external gamma dose (MARNA Project) (Sainz et al., 2014; Suárez et al., 1997, 2000; Quindós et al., 2004, 2008) and lithostratigraphic criteria. To decide the number of measurements per cell, it was essential to prioritise objectives and establish criteria. The decision on which criteria to use was made taking into account the objectives behind the European Radon map.

1. Surface criterion: The whole Spanish territory had to be covered by, at least one measurement per 10x10 km grid cell. 478 cells out of 5,478 total number of cells are inhabited and no data were collected. Thus, the lowest number of possible measurements was done over 5,000 cells.

2. Population criterion: In the first measurement campaign, an extra measurement was done for each town with a population exceeding 50,000 inhabitants, based on the Spanish National Statistics Institute (INE, 2014). Hence, an additional 1,000 measurements had to be made according to this criterion. In the second campaign this criterion was expanded to require a minimum of 6 measurements in cells including towns with populations larger than 200,000. A further 123 measurements were taken to meet this criterion.

3. MARNA criterion: Considering the importance of the geological factor it was decided to increase the number of measurements in areas with high radon potential. The starting point was the MARNA project (Suárez et al., 2000). MARNA determines potential radon emissions by taking into account the correlation between ^{226}Ra concentration in the soil and outdoor gamma dose levels. This criterion is based from 7,400 locations where measurements were performed to determine the potential for radon emission. These locations were linked to the 10x10km^2 cells, so-called MARNA cells. The different levels of exposure assigned to each cell correspond to the town with the higher dose. During the first campaign, additional measurements were taken in each cell identified in the MARNA project with a gamma exposure level over 35 nGy/h [4 μR/h]. 2 additional measurements were added for each location that exceeded 35 nGy/h [4 μR/h].

By doing so, 2,000 additional measurements were made (Sainz et al., 2014). Throughout the second campaign, measurements focused on towns with a gamma exposure level between 65 and 122 nGy/h [7.5 and 14 μR/h] (median risk) and involved a minimum of 6 measurements for each cell with this dose criterion. In the whole territory, 1,655 MARNA cells of this type were found, but the number of measurements taken was reduced to 960 due to limitations in budget.

The towns were classified according to their highest dose starting from 122 nGy/h and arranged in a decreasing order. A selection was made from the highest doses adjusted to the requirement of 6 measurements per cell, obtaining 960 measures. The result was the implementation of the measures in cells between 104 and 122 nGy/h [12 and 14 μR/h].

4. Lithostratigraphic criterion: Numerous studies support the importance of soil permeability factor in determining the radon potential in buildings (Kemski et al., 2001). Permeability is part of the lithological nature of the rock. The proper way to map this feature is by means of the lithostratigraphic map. It represents the lithostratigraphy homogeneously grouped by similar levels mapping permeability values (Garcia et al.,2013b).

Following the European Commission's guidelines (JRC-EC, 2014; JRC_REM_EC, 2014; JRC-IES, 2012) towards designing a European wide geological radon map, a series of areas of interest were identified in accordance with lithostratigraphic units for different regions and with that a specification of the number of measurements to be taken per unit and town (see Tables 2 and 3). The grouping of lithostratigraphies into units and regions of interest was due to the need of complying with the European Commission's premises for accounting for the permeability of the soils upon which buildings are constructed (Tollefsen et al., 2014).

By using the lithostratigraphic map from Geological and Mining Institute of Spain (IGME, 2014a; IGME, 2014b) at 1:200,000 scale, which includes the permeability factor of lithological units, units of interest were identified and cross checked with the local and regional cartographic databases (IGN, 2014). This increased the number of towns where measurements were to be taken. In order to identify the cells affected by this criterion, the above mentioned towns were superimposed onto the 10x10 km cell base and an additional 270 measurements were done. Figures 2 and 3 show the implementation of the previous criteria during the two measuring campaigns.

2.3 Radon measurements

The measurements were carried out using CR-39 track-etch detectors over an exposure period of 3 to 6 months. Having identified the sample cells, houses were selected in different ways. Random locations within the cell were selected and local institutions and government agencies were approached by telephone in most cases.

Each detector was distributed together with installation instructions, a form to be filled up, an information CD, some explanatory letters and a postage-paid envelope for returns. The form included questions regarding the building's design, building materials and occupants' lifestyle patterns.

The radioactivity laboratory of University of Cantabria (LaRUC) is validated to carry out these types of measurements under the validation scheme designed by PHE (Public Health England, UK) every year. Additionally, the laboratory evaluates the quality of the detector calibration by means of inter-comparison exercises at national and international levels on a regular basis (Gutierrez-Villanueva, 2011, 2015).

The measurements took place in ground-level buildings following the protocol of placing the detector in the main room at a height between 1 and 2 m on a shelf or wardrobe separate from the wall, and always away from air currents and heat sources. Once the measurement had been taken, a report was sent to the collaborator informing of the radon level in the buildings and including recommendations in order to reduce the concentration if it were deemed high.

2.4 Data representation

The measurement data were stored using the Geographic Information System's database including: identifying code of each detector; the result of the measurement (in Bq/m^3); location where the measurement was made; "x" and "y" coordinates and the cell of the location; and detector exposure time. Thence, the following data were obtained for each cell: number of measurements; geometric mean; arithmetic mean; median; minimum and maximum values and standard deviations. Their graphic representation was achieved by assigning these data to the "x" and "y" coordinates of the centroids of each cell. By doing so we ensured the anonymity of the project's collaborators, as the original data regarding the exact locations of the buildings were not shown on the map (Figure 4).

3. RESULTS

The Spanish indoor radon map now includes a total number of 9,211 measurements distributed across the country. The data conform to a log-normal distribution (Figure 5), as is commonly found when dealing with radon. The geometric mean and geometric standard deviation are the parameters often used to characterize this type of distribution.

The final result is the map shown in Figure 6. The cells with 1 measure were represented by the value of radon concentration obtained. The cells with a number of measurements between 2 and 5 were represented by using the arithmetic mean, and the cells with more than 6 radon measurements used the geometric mean.

The classification was carried out into four categories: < 50, 50-100, 100-300 and >300 Bq/m^3, in compliance with recommendations of the World Health Organization (WHO, 2009), the International Commission on Radiological Protection (ICRP, 2009, 2014) and the Nuclear Safety Council (CSN, 2012a) which establish the reference levels in the range 100 and 300 Bq/m^3. Among the Spanish data, 12% of the cells were in this range and 1% had geometric means higher than 300 Bq/m^3. This means that a large proportion of Spain's surface surpasses the limits recommended by international institutions and the national authority (table 4)

The majority of the Spanish territory is now covered by at least one measurement (Figure 8 and table 5), but there are areas with few measurements (between 1 and 5 measures, 52% of the surface). Incomplete cells (41%) are the result of not being able to carry out a complete analysis in successive sampling campaigns (inside this category 6 % of the cells is included without population). Whereas 7% of the surface has had greater than 6 measurements. This density of measures by cell is explained in the own criteria of sampling of the project.

Given that a part of the national territory is covered by only one measure, singular points with high concentration have been found in non prone areas. The significance of the values associated to these cells will increase with a higher number of measurements in future campaigns.

Table 6 shows a summary of the overall statistics of the data. As we can see, in comparison with previous measurement campaigns (CSN, 1998; Martín Matarranz, J.L, 2004; García et al., 2013a; Sainz et al., 2014), both the national geometric and the arithmetic mean have increased. This is due to the fact that in this second phase of the project the focus has been upon areas expected to have high radon exposure.

An interesting aspect is that areas with a high radon exposure were selected in accordance with lithostratigraphic criterion instead of lithologic or geologic criteria. The first criterion takes into account the soil permeability. Lithological maps show the most representative lithologic associations and geological maps show chronolithostratigraphic units (IGME, 2014b). The use of the lithostratigraphic map is justified by the fact that it incorporates the lithologic permeability factor, which is of great interest when analysing the behaviour of radon. Additionally, it represents units in a smaller scale and, consequently, in greater detail than the geologic or lithologic maps used up until now.

Previous studies have use lithostratigraphy to identify high radon areas in Spain (Garcia et al., 2013b). To test whether this criterion is valid or not, we investigated if it is possible to identify cells with radon concentrations higher than 300 Bq/m^3 and in the range 100-300 Bq/m^3 .

In order to verify the validity of lithostratigraphic criterion, 40 units identified as radon prone areas were selected and compared with available measurements in those areas. By overlapping the 42 cells over 300 Bq/m^3 with the above mentioned units, it was found that 88% of these cells were included within these areas. In the same way, the comparison of the 602 cells with a range of between 100 and 300 Bq/m^3, led to a 70% of agreement (see Figure 7 and Table 7).

The reason to find cells out of the mentioned comparison might be due to the different factors involved with high radon concentrations indoors (building materials, geological faults, ventilation rates, etc.). So it is necessary to perform extra measurements in those cells located in radon prone areas to validate the preliminary results. This would confirm the importance of lithostratigraphy to identify areas with high radon levels.

4. CONCLUSIONS

We have used different approaches to find areas with high radon concentrations: lithostratigraphy, lithology and geology. We conclude that the lithostratigraphy is most useful since it includes the soil permeability as a parameter, which is one of the keys to explain the radon transport in the soil. However, our study revealed that some of the high radon zones cannot be related to lithostratigraphical units. Therefore we need to increase the effort on gathering more radon data in order to generate a map that shows homogeneous geological units that may be identified as potential radon risk areas. The lithostratigraphical classification is sufficient to a national scale but questionable when we want to go into more local scales.

Despite the fact that the number of measurements in Spain has increased with the latest sampling campaigns, we have realised that that there are areas where the number of measurements remains low. Future campaigns should be focus on those areas with not enough radon measurements, as well as in those cells where high radon values have been found without correlation with lithostratigraphic classification.

We are going to develop probabilistic models of indoor radon presence to identify high radon risk areas on the basis of geologically similar units where the amount of measurements is lower. Also, in the future this model will enable us to verify the correlation between the different geographic variables in order to identify those which bring about the presence of higher concentrations to a greater extent.

5. ACKNOWLEDGEMENTS

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FIGURE CAPTIONS

Figure 1: Description of the sampling campaigns carried out towards the development of the Spanish indoor radon map

Figure 2: Schematic view of the criteria used in the first campaign resulting in 8,000 measurements

Figure 3: Extension of measurements to create the Spanish indoor radon map by including additional criteria: surface, lithostratigraphy and MARNA

Figure 4: Process used to incorporate the results of the measurements into the Geographic Information System (GIS)

Figure 5: Log-normal distribution of the current data included in the Spanish indoor radon map. Values of geometric mean (GM) and arithmetic mean (AM) are expressed in Bq/m^3.

Figure 6: The Spanish indoor radon map up-to-date. The cells include a total number of 9,211 radon determinations.

Figure 7: Lithoestratigraphic units identified as radon prone areas and cells with high radon concentration

Figure 8: Graphical view of the situation of the cells in terms of the number of data included in each cell.

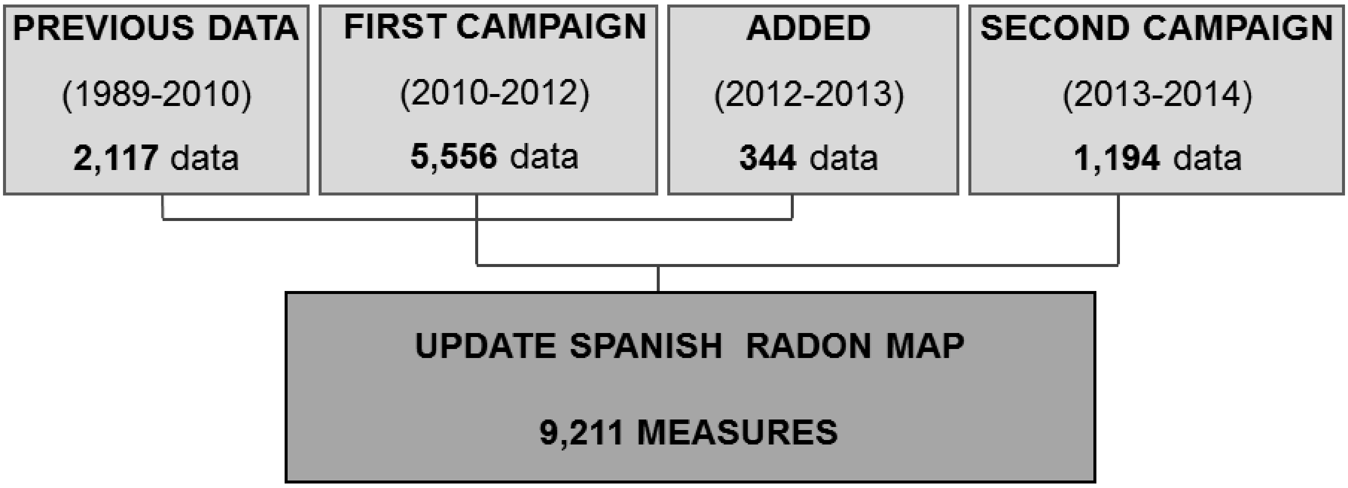

PREVIOUS DATA
(1989-2010)
2,117 data
FIRST CAMPAIGN
(2010-2012)
5,556 data
ADDED
(2012-2013)
344 data
SECOND CAMPAIGN
(2013-2014)
1,194 data
UPDATE SPANISH RADON MAP
9,211 MEASURES

Figure 1

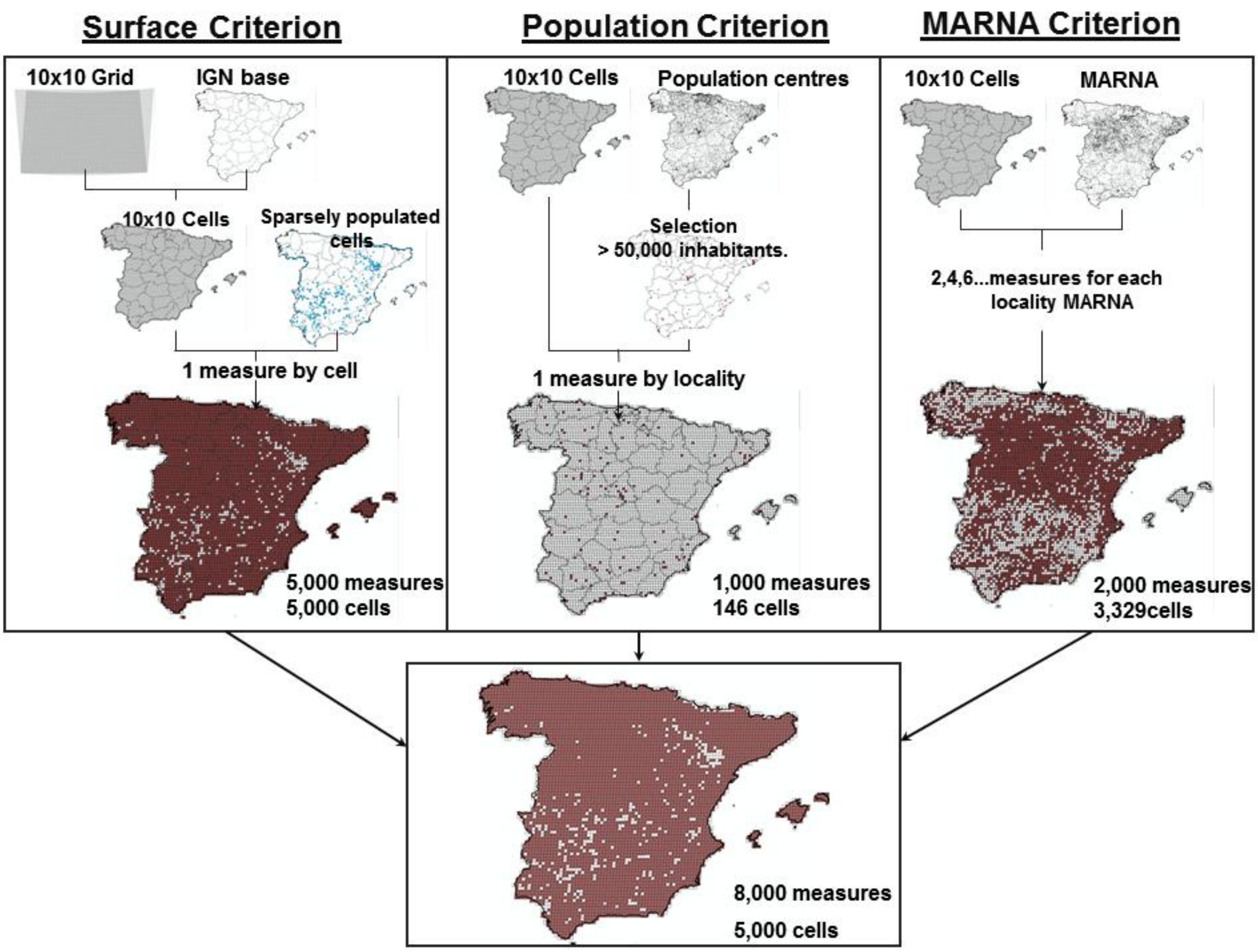
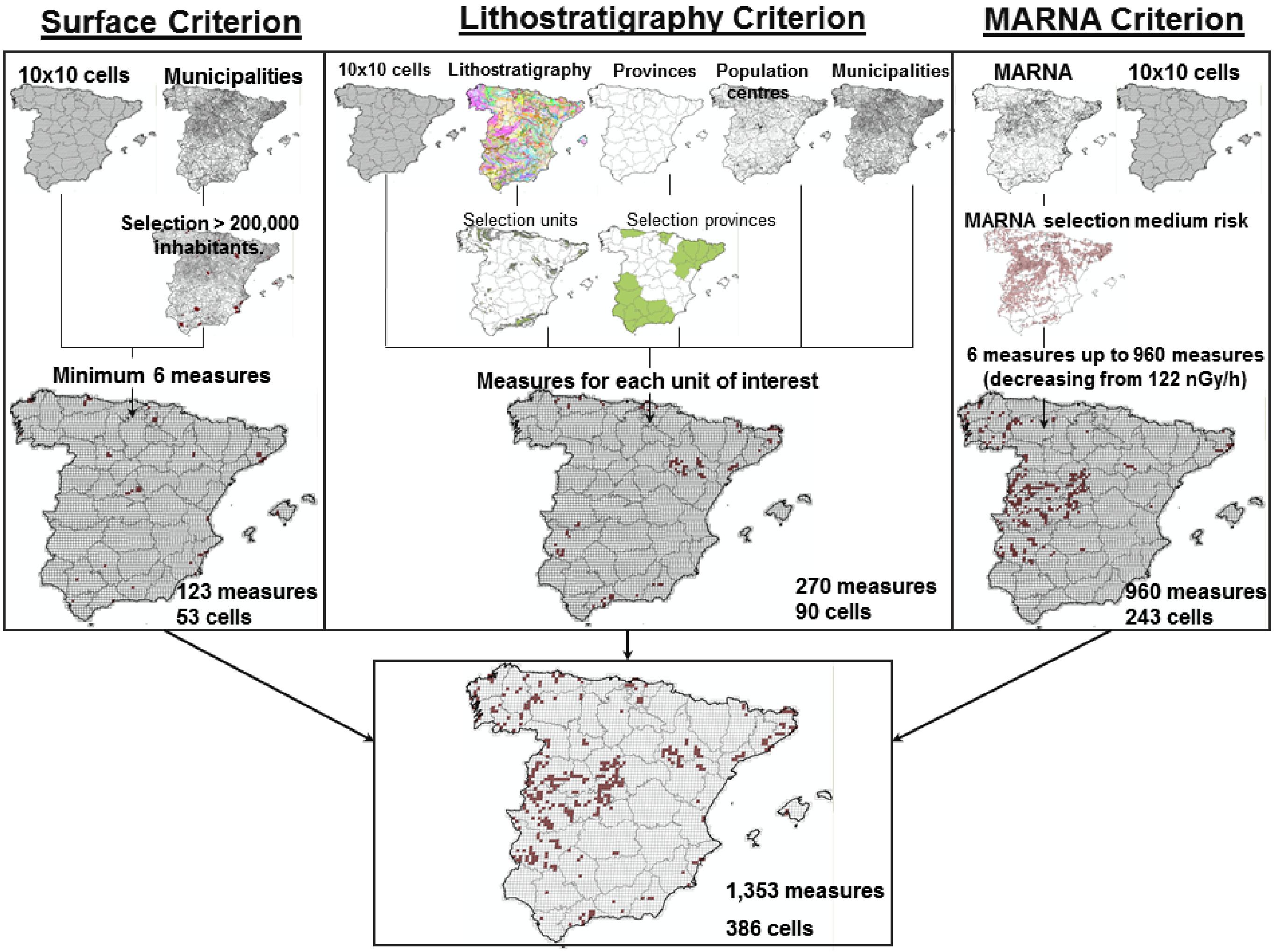

Surface Criterion
Population Criterion
MARNA Criterion
10x10 Grid
IGN base
10x10 Cells
Sparsely populated cells
1 measure by cell
5,000 measures
5,000 cells
10x10 Cells
Population centres
Selection
> 50,000 inhabitants.
1 measure by locality
1,000 measures
146 cells
10x10 Cells
MARNA
2,4,6...measures for each locality MARNA
2,000 measures
3,329cells
8,000 measures
5,000 cells

Figure 2



Surface Criterion
Lithostratigraphy Criterion
MARNA Criterion
10x10 cells
Municipalities
Selection > 200,000 inhabitants.
Minimum 6 measures
123 measures
53 cells
10x10 cells
Lithostratigraphy
Provinces
Population centres
Municipalities
Selection units
Selection provinces
Measures for each unit of interest
270 measures
90 cells
MARNA
10x10 cells
MARNA selection medium risk
6 measures up to 960 measures
(decreasing from 122 nGy/h)
960 measures
243 cells
1,353 measures
386 cells

Figure 3

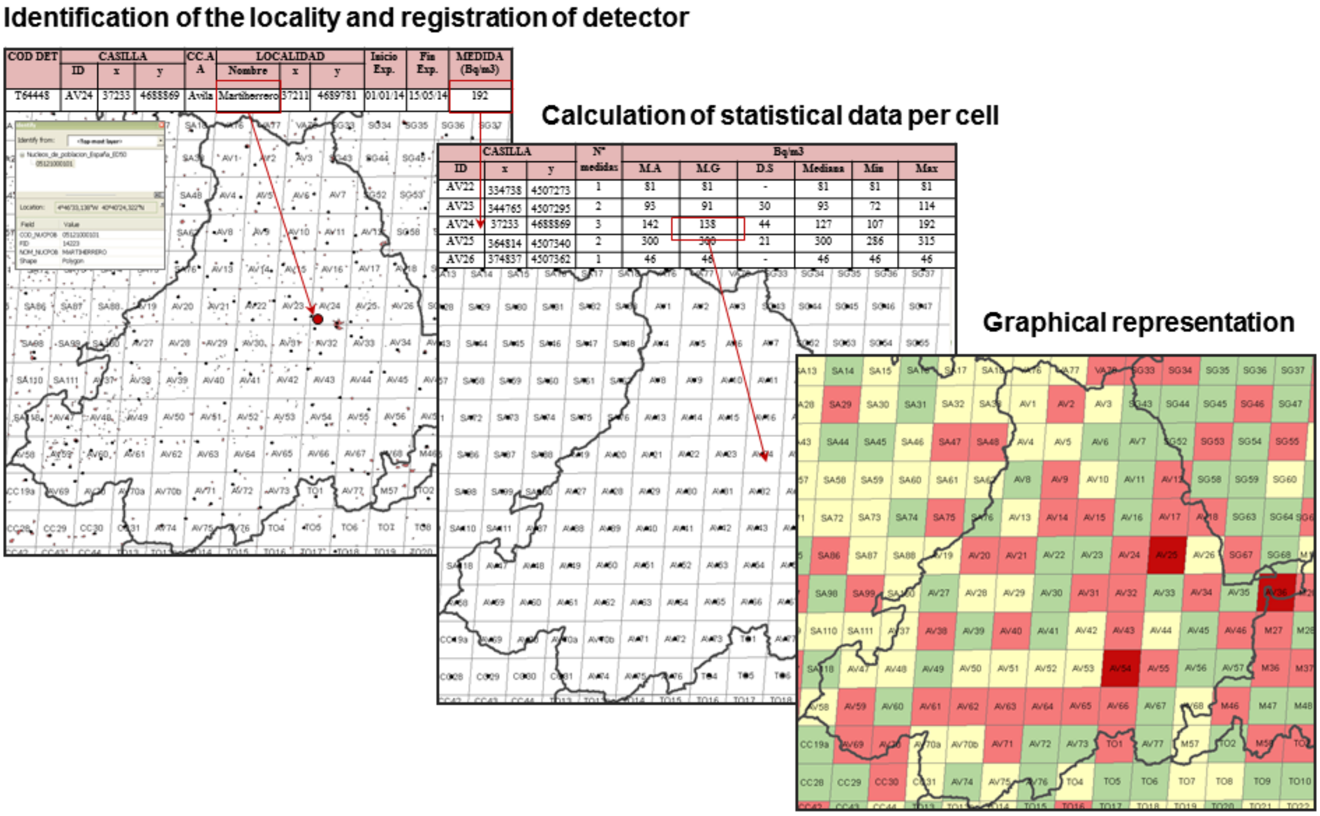
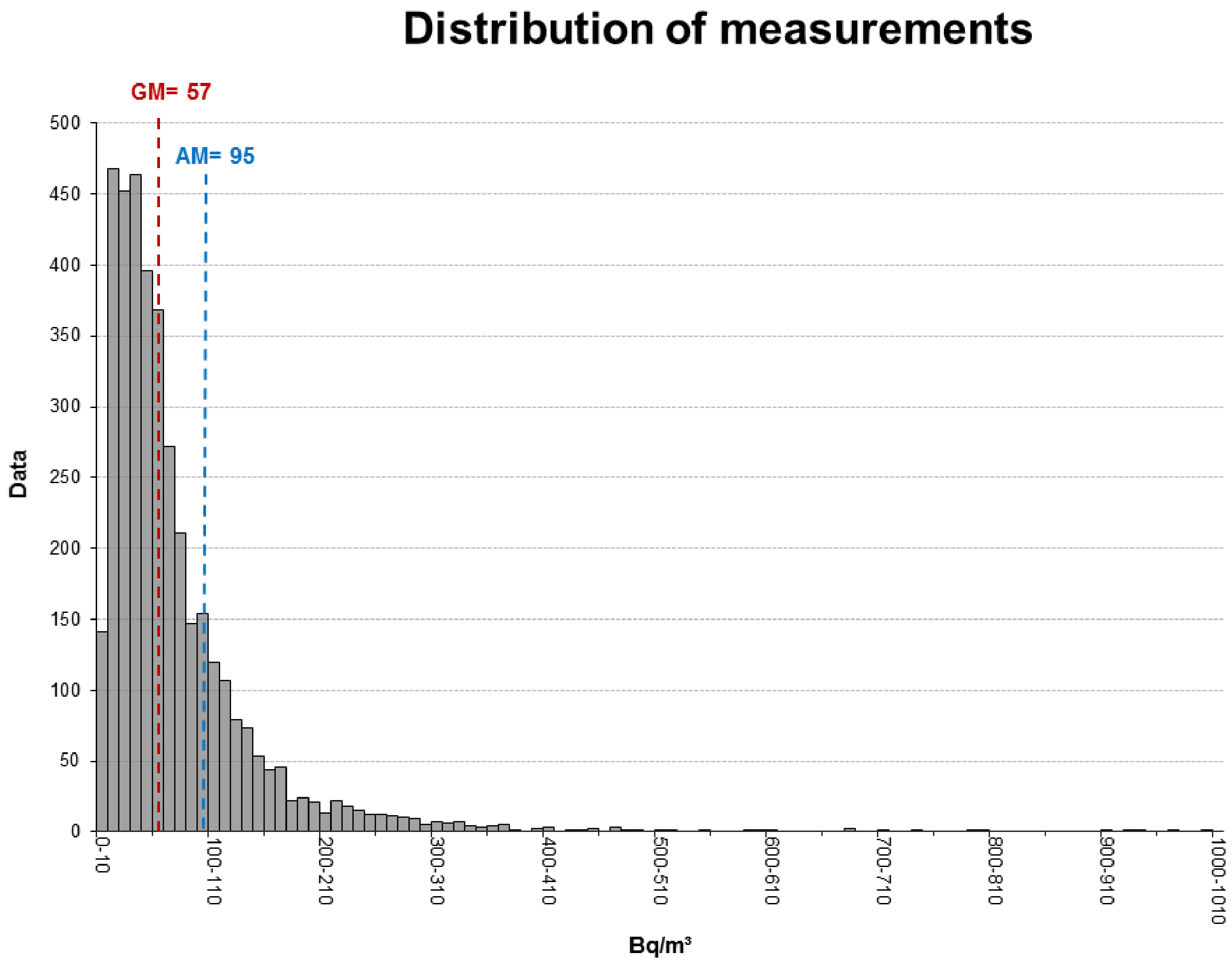

Identification of the locality and registration of detector
Calculation of statistical data per cell
Graphical representation

Figure 4



Distribution of measurements
GM= 57
AM= 95
Data
500
450
400
350
300
250
200
150
100
50
0
0-10
100-110
200-210
300-310
400-410
500-510
600-610
700-710
800-810
900-910
1000-1010
Bq/m³

Figure 5

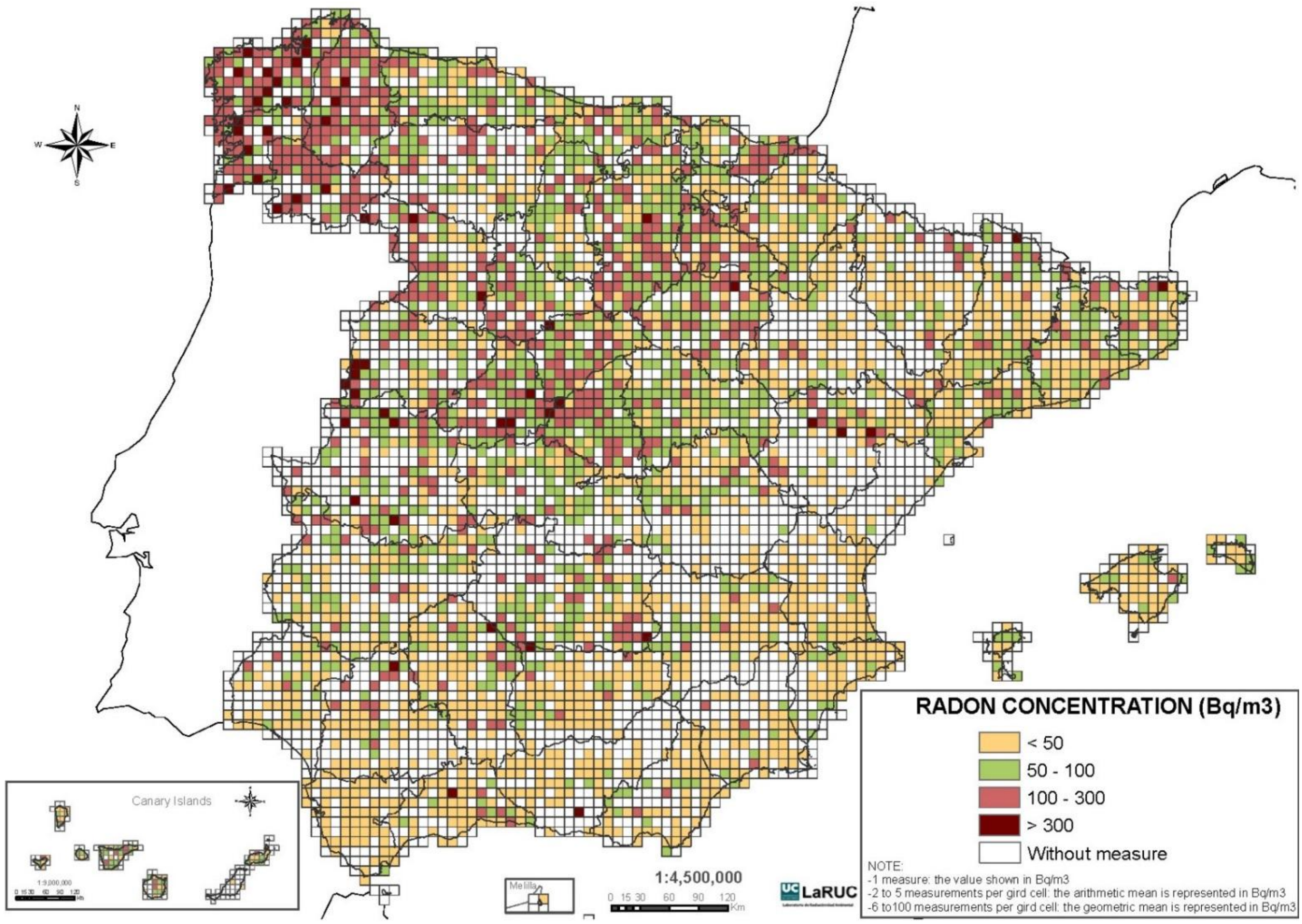

RADON CONCENTRATION (Bq/m3)
< 50
50 - 100
100 - 300
> 300
Without measure
NOTE:
-1 measure: the value shown in Bq/m3
-2 to 5 measurements per gird cell: the arithmetic mean is represented in Bq/m3
-6 to100 measurements per gird cell: the geometric mean is represented in Bq/m3
Canary Islands
Melilla
1:4,500,000
0 15 30 60 90 120
Km
N
S
W
E
LaRUC

Figure 6

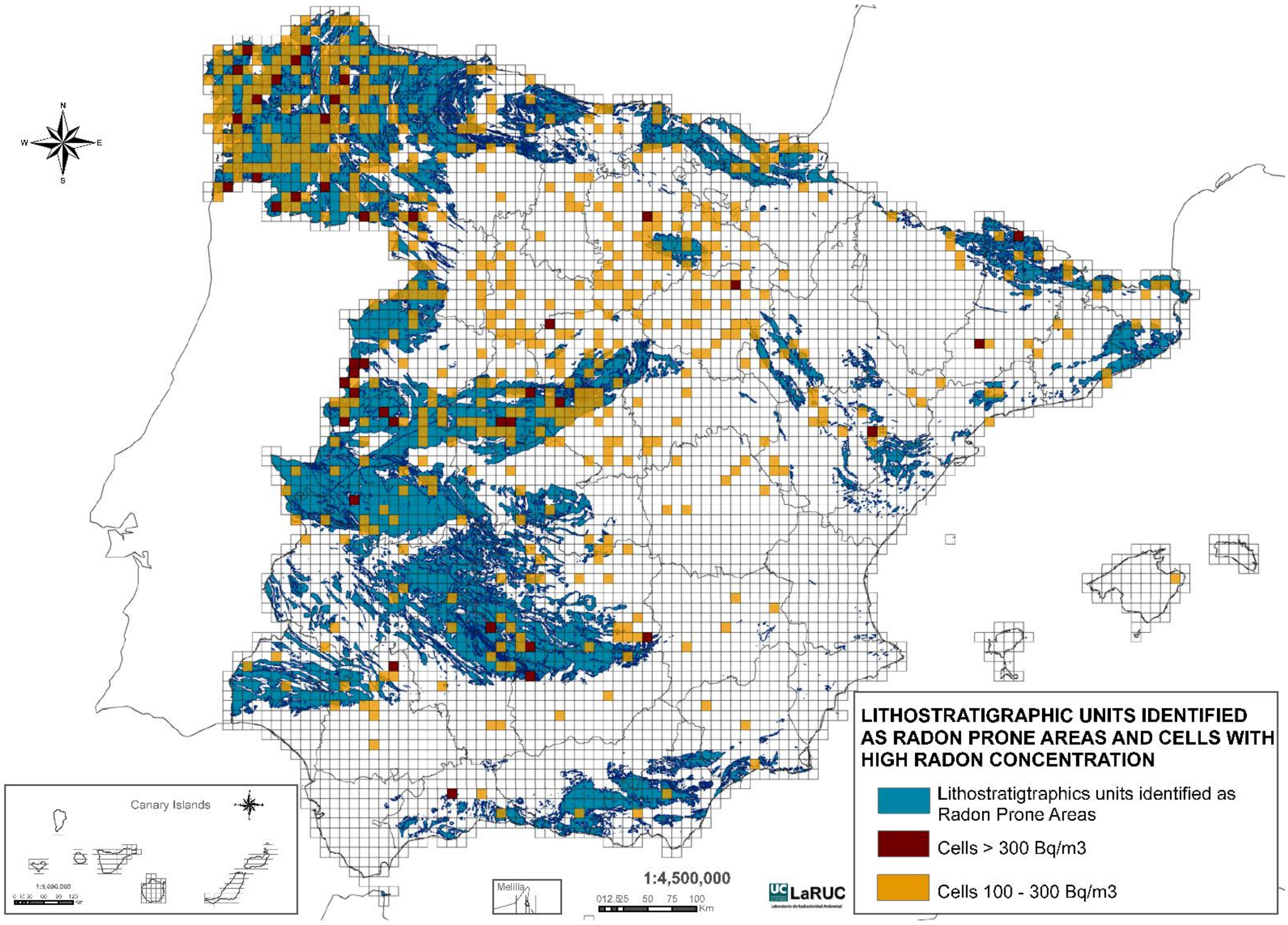
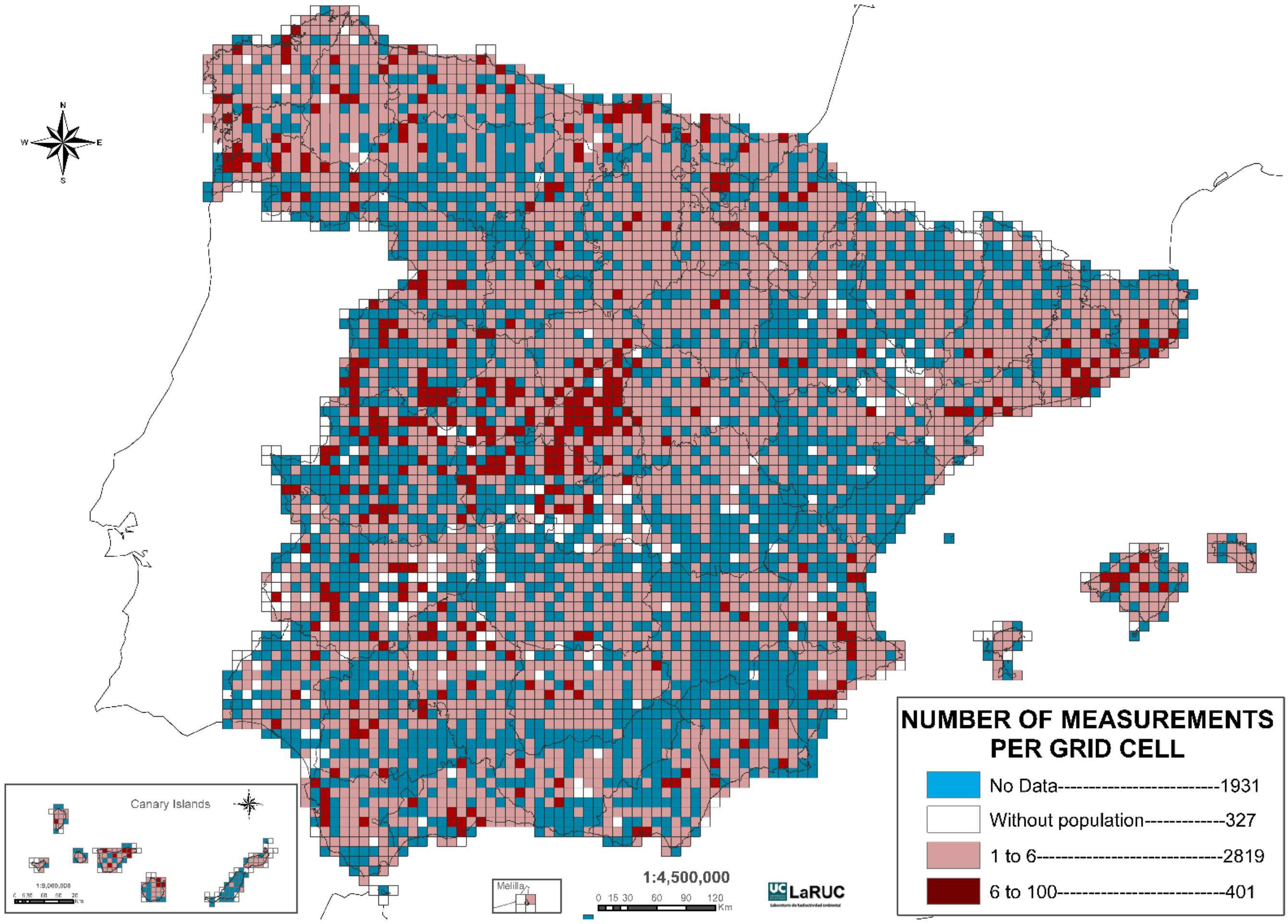

LITHOSTRATIGRAPHIC UNITS IDENTIFIED AS RADON PRONE AREAS AND CELLS WITH HIGH RADON CONCENTRATION
Lithostratigtraphics units identified as Radon Prone Areas
Cells > 300 Bq/m3
Cells 100 - 300 Bq/m3
Canary Islands
Melilla
1:4,500,000
Km
LaRUC

Figure 7



N
S
W
E
Canary Islands
1:9,000,000
Melilla
1:4,500,000
0 15 30 60 90 120
Km
LaRUC
NUMBER OF MEASUREMENTS PER GRID CELL
No Data--------------------------1931
Without population-------------327
1 to 6------------------------------2819
6 to 100---------------------------401

Figure 8

Table 1: Reference systems using in the project: ED50 UTM Zone 30N and defined by the European Commission (GISCO-LAEA)

	Reference System ED 1950 UTM Zone 30 N	**Reference System GISCO-LAEA**
Projection	Transverse Mercator	Lambert_Azimuthal_Equal_Area
False Easting	500000,000000	0,000000
False_Northing	0,000000	0,000000
Central_Meridian	-3,000000	9,000000
Scale_Factor	0,999600	
Latitude_Of_Origin	0,000000	48,000000
Linear Unit	Meter	Meter
GCS	GCS_European_1950	GCS_ETRS_1989
Angular Unit	Degree (0,0174532925199433299)	Degree (0,0174532925199433299)
Prime Meridian	Greenwich (0,000000000000000)	Greenwich (0,0000000000000000)
Spheroid	International_1924	GRS_1980
Semimajor Axis	6378388,0000000000000000	6378137,0000000000000000
Semiminor Axis	6356911,9461279465000000	6356752,3141403561000000
Inverse Flattening	297,0000000000000000	298,2572221010000200000

Table 2: Description of the spatial units of interest as used in this work and their lithostratigraphical definition.

Lithostratigraphic unit code (IGME)	**Lithostratigraphy definition**
130	Limestones, calcoschists and whiteboards
131	Micaschists, gneises, phyllites, quartzites and plasters
133	Micaschists, quartz and gneises
79	Slates, sandstone and quartzite. Series of los Cabos
85	Slates, sandstones and microconglomerates. Slates from Lancea
86	Quartzites, shales and rocks volcanocl. and volcanosed. Quartzite from Barrios and FM Oville
91	Slates and sandstones. Huergas slates
104	Quartzites, slates, sandstones, shales, limestones and dolomites. Paleozoic Iberian Aragon
117	Quartzites and slates
369	Shales with interbedded carbonate and gypsum
7	Plutonic basic Hercinian rocks (gabbros, dioritas, tonalite, ultramafic rocks)
2	Acid rocks metamorphic (Otogneises,migmatitas). Gn. gland., metarriolitas (Ollo Sapo).
127	Phyllites, schists, quartzites, limestones, slates and corneal (metamorphic)
152	Sandstones, sands, sandy limestones, marls, clay and loamy
153	Sandstones, shales and marls

173	Limestone reef, rudistas, bioclastic limestones, dolomites and marl
174	Marl, limestone, clay and dolomites

Table 3: Examples of the unit numbers of interest in some Spanish regions and the number of measurements necessary to take. Not all Spanish regions are represented.

Region	**Lithostratigraphic unit code of interest (IGME)**	**Measures to take**	**Criterion**
Andalucía	130-131-133	40	In at least 6 different municipalities (covering 3 units) with a minimum of 6 measures by municipality
Asturias	79-85-86-91	50	In at least 8 different municipalities (covering 4 units) with a minimum of 6 measures by municipality
Aragón	104-117-369	40	In at least 6 different municipalities (covering 3 units) with a minimum of 6 measures by municipality
Extremadura	7	30	In at least 4 different municipalities with a minimum of 6 measures by municipality
Cataluña	2-7-117-127	60	At least 3 municipalities in unit 2, 3 municipalities in unit 7 and 3 to 117 and 127 units. With a minimum of 6 measures by municipality
País Vasco	152-153-173-174	50	In at least 8 different municipalities (covering 4 units) with a minimum of 6 measures by municipality

Table 4: Concentration of radon: Number of cells and data

Range (Bq/m^3)	Number of cells	Number of data
< 50	1606 (29%)	4294 (46%)
50 - 100	967 (18%)	2922 (32%)
100 - 300	602 (11%)	1902 (21%)
> 300	42 (1%)	93 (1%)

Table 5: Number of measures per cell

Number of measures (Per cell)	**Number of cells** (%)
No data	41
1	29
2	12
3	6
4	3
5	2
6 to 100	7

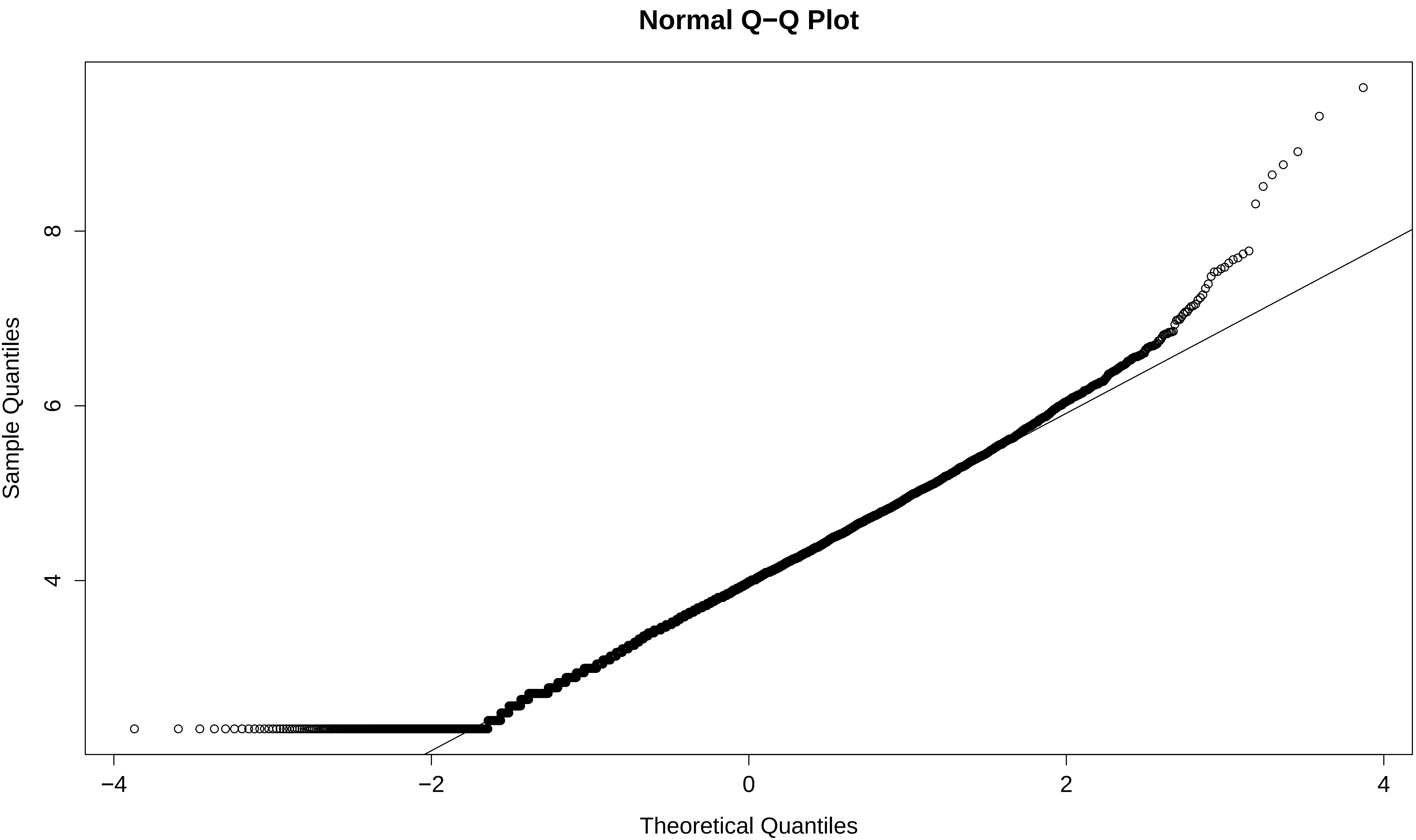
Table 6: Descriptive statistics of data used to produce the Spanish indoor radon map up-to-date. (Bq/m^3)

Number of measurements	Arithmetic Mean	Arithmetic Standard Deviation	Geometric Mean	Geometric Standard Deviation	Median	**RANGE**
9,211	95.0	270	56.6	2.6	54	10-15,400

Table 7: Comparison on the effectiveness of different approaches to identify high radon risk areas.

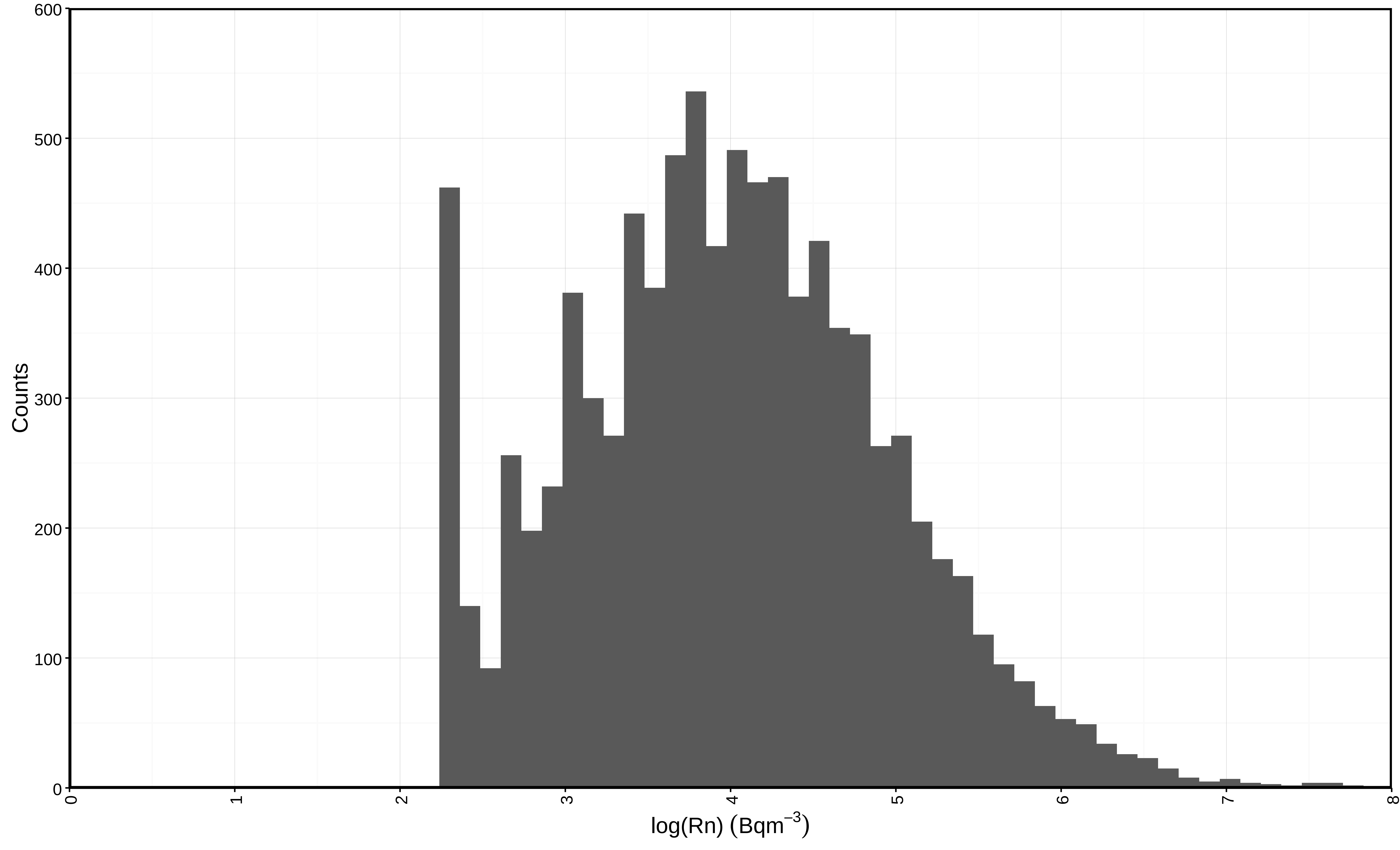
	Lithostratigraphy Scale 1:200,000	**Lithology Scale 1:1,000,000**	**Geology Scale 1:1,000,000**
All units	329	26	16,041
Units within radon prone areas	40	20	7,236
Number of cell inside radon prone areas	>300 Bq/m^3 37(88%) and 100-300 Bq/m^3 421(70%)		

Histogram


Normal Q−Q Plot
Sample Quantiles
Theoretical Quantiles
4
6
8
−4
−2
0
2
4

QQ-PLOT

Click here to download Supplementary Material: 20160420_rsw_qqplot (1).pdf



Counts
log(Rn) (Bqm^{-3})
0
100
200
300
400
500
600
0
1
2
3
4
5
6
7
8